

## Plastic Part Comprising Lustrous Pigments And Filler Particles

5 The invention relates to a plastic part which may comprise lustrous pigments and filler particles and optionally assistants for plastics processing, such as one or more pigments, stabilisers, plasticisers, fillers and/or reinforcing materials. Plastic parts of this type exhibit a pronounced glitter effect.

10 The term "lustrous pigments" is defined as pigments in which lustrous effects arise due to directional reflections at metallic or strongly light-refracting pigment particles which have a predominantly two-dimensional shape and alignment. These include pearlescent pigments, which are lustrous pigments which comprise colorless, transparent and highly light-refracting platelets. Due to parallel alignment of pearlescent pigments in  
15 plastics or in surface coatings, multiple reflections cause a soft lustrous effect which is known as pearlescence. Pearlescent pigments having a certain layer thickness may satisfy the interference conditions and then exhibit iridescent colors, thus giving rise to the name "interference pigments". The hue here is dependent on the viewing angle. A further  
20 feature of these transparent interference pigments is that complementary colors are visible in reflected light and transmitted light. In addition, there are also interference pigments with non-transparent support materials in which only the color in reflected light is visible. Lustrous pigments include nacreous pigments.

25 The lustrous pigments further include the commercially available metal-effect pigments, such as, for example, aluminium platelets, the goniochromatic lustrous pigments based on silica platelets, aluminium platelets or iron oxide platelets as support material and the liquid-crystal  
30 pigments.

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Lustrous pigments must have a certain particle size in order to provide a glitter effect to the eye. The eye is capable of recognising an individual pigment platelet in this case as a specific particle. Since the resolving power of the eye is greatly affected by the contrast of the structures to be resolved, a specific particle size at which a glitter effect arises cannot be quoted. In general, lustrous pigments having a particle size of less than 25  $\mu\text{m}$  give rise to a silk to satin gloss. At particle sizes up to about 60  $\mu\text{m}$ , a slightly structured sheen is observed, but it is not yet possible to refer to glittering. Glitter effects are observed at particle sizes of 80 to 100  $\mu\text{m}$ . However, particles of this size cannot always be produced, for example, single crystals of basic lead carbonate or  $\text{BiOCl}$  or  $\text{Al}_2\text{O}_3$  platelets as substrates for layer/substrate pigments can only be produced with difficulty in sizes of this order. In the case of mica pigments, the production of relatively large particles is less problematic, and pigments of this type are also commercially available.

The known lustrous pigments, including the pearlescent pigments, give rise to lustrous effects, but not glitter effects, in application systems, for example, plastics, owing to their limited particle size.

An object of the invention is to provide plastic parts which comprise lustrous pigments which provide a glitter effect or reinforce the glitter effect in pigments which already exhibit this effect.

Upon further study of the specification and appended claims, further objects and advantages of this invention will become apparent to those skilled in the art.

These and other objects of the invention are achieved by adding filler particles which have a substantially isometric body shape to the plastic, in addition to the lustrous pigment, before processing, where the diameter of these filler particles is in the range of 10 to 200  $\mu\text{m}$ . In addition, assistants

in plastics processing, some of which are described in detail in  
"Taschenbuch der Kunststoff-Additive; Stabilisatoren, Hilfsstoffe,  
Weichmacher, Füllstoffe, Verstärkungsmittel, Farbmittel" [Handbook of  
Plastics Additives; Stabilisers, Assistants, Plasticisers, Fillers, Reinforcing  
Materials and Colorants], 2nd Edition, Carl Hauser Verlag, Munich, Vienna,  
may also be present.

The invention thus relates to a plastic part comprising lustrous pigments  
comprising metallic or strongly light-refracting pigment particles or platelets  
which have a predominantly two-dimensional shape and are aligned  
substantially parallel with the surface of the plastic part, and filler particles  
comprising hollow, solid or compact particles which have a substantially  
isometric body shape and have a diameter of 10 to 200  $\mu\text{m}$ . Two-  
dimensional refers to particles, which are actually three-dimensional, but  
the thickness is small.

The filler particles preferably are small transparent beads, which are  
hollow, solid or compact. Deviations from the ideal spherical shape are  
possible, but the filler particles should predominantly have virtually  
isometric shapes. This means that the dimensions of the particles are  
approximately the same in all three spatial directions. The surface of the  
filler particles is preferably smooth. In the case of rough surfaces, for  
example, in ground filler particles, although a coarse glitter effect is  
observed, the colors have, for example, somewhat reduced brilliance.  
Preferred fillers are glass beads or hollow glass beads from the Minnesota  
Mining and Manufacturing Company. Materials, other than glass may be  
used, such as various duroplastics which do not melt or dilute during the  
production of the plastic part, or  $\text{SiO}_2$  spheres. The refractive index of the  
filler particles material should be approximately in the order of the refractive  
index of the matrix material into which the filler particles and lustrous  
pigments are incorporated in order to avoid excessively impairing the  
transparency and thus the lustrous of the pigmented matrix material. The

shape and surface area of the filler particles are of importance. Of lesser importance is the material of which the filler particles are made. Additionally, the filler particles should not melt or dilute in the matrix material during the production of the plastic part.

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An important parameter when selecting the filler particles is the diameter of the filler particles. This is between 10 and 200  $\mu\text{m}$ , with preferred particle diameters being between 15 and 150  $\mu\text{m}$ , in particular between 20 and 120  $\mu\text{m}$ . The amount of filler particles is from 0.2 to 10 parts by weight, preferably from 0.5 to 5 parts by weight and in particular from 0.5 to 3 parts by weight, based on the total weight of the plastic part.

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Lustrous pigments which can be employed are platelet-shaped pigments, such as metal-effect pigments, goniochromatic lustrous pigments, interference pigments, pearlescent pigments and liquid-crystal pigments.

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Metal-effect pigments comprise platelets of the metals aluminium, copper, zinc, tin and alloys thereof, in particular aluminium and gold-bronze alloys. The surface of the metal platelets can be passivated or provided with a protective layer, for example, of metal oxides. Metal-effect pigments are marketed by the Eckart company under the trade name Standard®.

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Goniochromatic lustrous pigments comprise multicoated platelet-shaped metallic substrates, transparent non-metallic substrates or multicoated metal-oxide platelets. Aluminium platelets are employed as metallic substrate, mica is employed as transparent non-metallic substrate and iron oxide is employed as metal-oxide platelets. These lustrous pigments are described in greater detail in EP 741 170, EP 708 154 and EP 753 545. They are available from BASF under the trade names Paliochrom® and Variochrom®.

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Multilayered interference pigments having a light-opaque aluminium layer as the central layer are produced by the Flex company under the trade names Chromaflair® and OVP® (optically variable pigments). These pigments, which are primarily employed in security printing, basically have a five-layered structure. On a central light-opaque aluminium layer, layers of magnesium fluoride as interlayers and subsequently semi-transparent chromium layers as outer layers are deposited on both sides. The pigments are described in US 4,434,010.

Interference pigments having transparent support materials are known as pearlescent pigments. The platelet-shaped transparent support materials may be mica, other phyllosilicates, such as talc or kaolin, glass flakes, SiO<sub>2</sub> flakes, TiO<sub>2</sub> flakes or Al<sub>2</sub>O<sub>3</sub> flakes. These support materials are coated with one or more metal-oxide layers. The metal oxides used here are both colorless high-refractive-index metal oxides, such as, for example, titanium dioxide or zirconium oxide, colorless low-refractive-index metal oxides, such as, for example, silicon dioxide or aluminium oxide, and colored metal oxides, such as, for example, chromium oxide, cobalt oxide and in particular iron oxides. These platelet-shaped pearlescent pigments are known and for the most part commercially available. Pearlescent pigments are described, for example, in German Patents 31 37 808, 31 37 809, 31 51 343, 31 51 354, 31 51 355, 32 11 602 and 32 35 017. These pigments are available from Merck KGaA under the trade names Iriodin®, Colorstream® and Xirallic®.

Liquid-crystal pigments are interference pigments based on liquid-crystalline polymers. The individual pigment particles are fragments of a thin crosslinked film of liquid-crystalline polymers. The color effects which can be achieved therewith are based on the regular structure and homogeneous arrangement of the molecules in the form of a liquid crystal and on interference, attributable thereto, of a certain spectral light fraction which is reflected by the pigment. The other light fractions pass through the

pigment. Liquid-crystalline interference pigments are described in US 5,807,497 and US 5,824,733. They are available from Wacker-Chemie GmbH under the trade name HELICONE® HC.

5 The diameter of the particles of the non-glittering lustrous pigments employed in accordance with the invention is in the range of 2 to 80  $\mu\text{m}$ . The pigments can be used individually or in the form of a pigment mixture. Pigment and filler particles can likewise be added as individual components or in the form of a mixture to the matrix material, for example plastic  
10 granules. The sequence of addition of the two components is unimportant.

Lustrous pigments are generally employed in plastics in concentrations of 0.5-2% by weight. In films or thin layers, significantly higher concentrations are necessary. The crucial factor for the pigment concentration needed is  
15 the desired effect.

The addition of filler particles to the lustrous pigments causes the latter to be deflected out of their more or less ideal alignment parallel to the surface. A certain tilt of the pigments toward one another occurs, i.e., they are no  
20 longer aligned substantially parallel with the surface of the plastic part, giving rise to the visual impression of individual particles having larger particle diameters than the actual particle diameters. This visual impression results in an apparent coarsening of the surface of the pigment particles tilted toward one another, giving rise to a glitter effect which is observable  
25 over a much greater viewing-angle range than a glitter effect which can be achieved with lustrous pigments having larger particle sizes.

In addition, the effects of the pigments which are larger than 80  $\mu\text{m}$  are also influenced, i.e., reinforced. In the case of lustrous pigments which  
30 already exhibit a glitter effect owing to their particle size, the glitter effect is reinforced and becomes visible over a greater angle range.

The use of the filler particles together with strongly changing interference pigments, available, for example, under the trade name "Colorstream®", produced by Merck KGaA, reduces the angle dependence of the color change (flop). The transition from one color to another color becomes softer, i.e. the angle range within which the color changes becomes greater.

The use of filler particles in the plastic part may gradually reduce the visibility of flow lines and weld lines.

For a plastic part according to the invention, use can be made of known, transparent plastics, in particular thermoplastics. Suitable plastics are, for example, polyethylene (PE), polypropylene (PP), polystyrene (PS), polyphenylene oxide, polyacetal, polybutylene terephthalate, polymethyl methacrylate, polyvinyl acetate, acrylonitrile-butadiene-styrene (ABS), acrylonitrile-styrene-acrylate (ASA), polycarbonate, polyether sulfone, polyether ketones and copolymers and/or mixtures thereof. In addition, it is also possible to use casting resins, for example unsaturated polyester and methyl methacrylate casting resins.

In the case of so-called direct coloring, the plastic granules are uniformly wetted at the surface with coupling agents, for example, diisooctyl phthalate, and lustrous pigments and filler particles are then added and distributed uniformly on the surface of the plastic granules by mixing. This mixture is then processed directly in an injection-molding machine. The moldings obtained exhibit a very homogeneous distribution of the lustrous pigments.

Direct coloring is a process which is preferably used in the laboratory. In production, masterbatches are employed. The term masterbatch is applied to a pigment preparation in which the pigment is in a significantly higher

concentration than in the end product and is in fully dispersed form in a matrix which is compatible with the starting material. The incorporation of lustrous pigments into plastics is described in detail in "Perlglanzpigmente" [Pearlescent Pigments], edited by Dr. Ulrich Zorll, Curt R. Vincentz Verlag, Hanover 1996.

Lustrous pigments and filler particles are incorporated into unsaturated plastics by stirring-in before the casting process.

Without further elaboration, it is believed that one skilled in the art can, using the preceding description, utilize the present invention to its fullest extent. The following preferred specific embodiments are, therefore, to be construed as merely illustrative, and not limitative of the remainder of the disclosure in any way whatsoever.

In the foregoing and in the following examples, all temperatures are set forth uncorrected in degrees Celsius; and, unless otherwise indicated, all parts and percentages are by weight.

The entire disclosures of all applications, patents and publications, cited above or below, and of corresponding German application No. 10106198.6, filed February 10, 2001, is hereby incorporated by reference.

## Examples

### Example 1

978 g of PP granules having the trade name Stamylin® PPH 10 from DSM are wetted on the surface with 2 g of diisooctyl phthalate in a tumble mixer and subsequently mixed well. 10 g of TiO<sub>2</sub>-coated mica pigments having a particle size of 5 to 25 µm from E. Merck, Darmstadt, Federal Republic of Germany (Iriodin 123 satin) and 10 g of filler particles are subsequently added and distributed uniformly on the surface of the plastic granules by



mixing. The filler particles are hollow glass beads having diameters of essentially 20 to 80  $\mu\text{m}$  which are available under the trade name Scotchlite® K37 from the 3M Company, USA. The plastic granules pigmented in this way are converted into moldings by injection molding by means of an injection-molding machine. Compared with Comparative Example 1, the finished moldings exhibit significantly more coarsely structured pearlescence which does not correspond to the usual appearance in accordance with the particle size of the pearlescent pigments employed. This more coarsely structured pearlescence represents a glitter effect. The angle dependence of the lustrous effect is less than in the comparative example. The proportion by weight of the pearlescent pigment and filler particles in the molding is in each case 1%.

### Example 2

The same components as in Example 1 are used, but the Scotchlite® K37 hollow glass beads are replaced by Scotchlite® K1 hollow glass beads from the 3M Company, USA. The diameter of the filler particles is essentially in the range of 30 to 110  $\mu\text{m}$ . The glitter effect observed visually corresponds to that in Example 1.

### Example 3

As in Example 1, 978 g of PP granules (Stamylan® PPH10 granules) are wetted on the surface with 2 g of diisooctyl phthalate in a tumble mixer and subsequently mixed well. 10 g of a silver-colored pearlescent pigment comprising  $\text{Al}_2\text{O}_3$  as substrate and a coating of  $\text{TiO}_2$  and 10 g of filler particles (Scotchlite® K37 hollow glass beads from the 3M Company, USA) are then added and distributed uniformly on the surface of the plastic granules by mixing. Pigments with  $\text{Al}_2\text{O}_3$  as substrate are available under the trade name Xirallic® from Merck KGaA. The plastic granules pigmented in this way are converted into moldings by injection molding by means of an injection-molding machine. The individual molding exhibits a significantly

coarser, structured pearlescence than is to be expected of the pigment used owing to its particle size. This pearlescence evokes the visual impression of a glitter effect.

5      **Example 4**

983 g of PP granules (Stamylan® PPH10) are wetted on the surface with 2 g of diisooctyl phthalate and subsequently mixed well. 5 g a changing pearlescent pigment comprising SiO<sub>2</sub> as substrate and Fe<sub>2</sub>O<sub>3</sub> as coating and 5 g of Scotchlite® K37 hollow glass beads from the 3M Company, USA, are then added and distributed uniformly on the surface of the plastic granules by mixing. The pearlescent pigments are available under the trade name Colorstream® from Merck KGaA, Darmstadt, Federal Republic of Germany. The proportion by weight of the pearlescent pigment and filler particles in the plastic part is in each case 0.5%. The pigmented plastic granules are converted into moldings by injection molding by means of an injection-molding machine. The injection moldings obtained exhibit a significantly coarser, structured pearlescence than would have been expected of this pigment on the basis of its particle size. The color transition from blue-red to greenish is softer and the color flop occurs in a broader viewing-angle range than in the case of use of the pigments without filler particles.

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**Comparative Example**

25      988 g of PP granules (available from DSM, the Netherlands, under the trade name Stamylan® PPH10) are wetted on the surface with 2 g of diisooctyl phthalate in a tumble mixer and subsequently mixed well. 10 g of lustrous pigments based on TiO<sub>2</sub>-coated mica pigments having a particle size of 5 to 25 µm are then added (commercial product Iridin® 123 Glanzsatin from Merck KGaA, Darmstadt, FRG) and distributed uniformly  
30      on the surface of the granules by mixing. These pigmented granules are converted into moldings by means of an injection-molding machine. The

lustrous pigment has a proportion by weight of 1% in the finished molding. The finished moldings exhibit good, not very structured pearlescence in accordance with their particle size.

5 The preceding examples can be repeated with similar success by substituting the generically or specifically described reactants and/or operating conditions of this invention for those used in the preceding examples.

10 From the foregoing description, one skilled in the art can easily ascertain the essential characteristics of this invention and, without departing from the spirit and scope thereof, can make various changes and modifications of the invention to adapt it to various usages and conditions.

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